Form PTO-1390 (Rev. 12-29-99) US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		ATTORNEY'S DOCKET NO **C 2041 PCT/US** U.S. APPLICATION NO. (if known see 37 CFR 1.5) **10/049961**
INTERNATIONAL APPLICATION NO. **PCT/EP00/07717**	INTERNATIONAL FILING DATE **August 9, 2000**	PRIORITY DATE CLAIMED **August 18, 1999**

TITLE OF INVENTION

DECORATIVE COSMETIC PREPARATIONS CONTAINING CHITOSAN MICROCAPSULES CHARGED WITH ACTIVE INGREDIENTS

APPLICANT(S) FOR DO/EO/US

Maria DeMORAGAS, Josep GARCES GARCES, Josep-Lluis VILADOT PETIT

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1. ■ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.

2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.

3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).

4. ■ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.

5. ■ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ■ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).

6. ■ A translation of the International Application into English (35 U.S.C. 371(c)(2)).

7. ■ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ■ have not been made and will not be made.

8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).

9. ■ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). **(UNEXECUTED)**

10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.

12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.

13. ■ A FIRST preliminary amendment
    ☐ A SECOND or SUBSEQUENT preliminary amendment.

14. ☐ A substitute specification.

15. ☐ A change of power of attorney and/or address letter.

16. ☐ Other items or information:

"Express Mail Post Office to Addressee" service Mailing Label Number EL541614404US .

JC13 Rec'd PCT/PTO 19 FEB 2002

U.S. Application No. (If known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
10/049961	PCT/EP00/07717	C 2041 PCT/US

17. ■ The following fees are submitted:

	CALCULATIONS	PTO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... **$1,040.00**		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... **$890.00**		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37CFR 1.445(a)(2)) paid to USPTO **$740.00**		
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) **$710.00**		
International preliminary examination fee paid to USPTO (37CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... **$100.00**		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 890	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).	$ 0	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total Claims	20 - 20 =	0	0 X $18.00	$ 0	
Independent Claims	2 - 3 =	0	0 X $84.00	$ 0	
Multiple dependent claims (s)(if applicable)		0	+ $280.00	$ 0	
TOTAL OF ABOVE CALCULATIONS =				$ 890	
Reduction of ½ for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				$ 0	
SUBTOTAL =				$ 890	
Processing fee of **$130.00** for furnishing the English translation later the ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +				$ 0	
TOTAL NATIONAL FEE =				$ 890	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property +				$ 0	
TOTAL FEES ENCLOSED =				$ 890	
				Amount to be: refunded:	$----------
				charged:	$890.00

a. ☐ A check in the amount of $_____ to cover the above fees is enclosed.

b. ■ Please charge my Deposit Account No. 50-1177 in the amount of $890.00 to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. 02-0077.

c. ■ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 50-1177. A triplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO: **Customer Label No. 23657**

SIGNATURE

Aaron R. Ettelman
NAME ATTORNEY FOR APPLICANT

42,516
REGISTRATION NUMBER

"Express Mail " Mailing Label Number EL541614404US .

PATENT
Docket No. C 2041 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE: PCT/EP00/07717
International Filing Date: August 9, 2000
Priority Date Claimed: August 18, 1999
Applicant: DeMoragas, et al.
Title: DECORATIVE COSMETIC PREPARATIONS CONTAINING CHITOSAN MICROCAPSULES CHARGED WITH ACTIVE INGREDIENTS
Applicants' Reference: C 2041 PCT/US

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

Prior to the calculation of fees and examination of the above-identified national stage application pursuant to the accompanying submission under 35 U.S.C. §371, please amend the English translation of the International Application submitted herewith, without prejudice, as follows:

In the Specification:

Please amend the instant Specification, without prejudice, as follows:

Please delete all text above line 9 of page 1, including the heading "Prior Art", and replace the deleted matter with the following new section headings and title of the invention:

--TITLE OF THE INVENTION

Cosmetic Preparations Containing Chitosan Microcapsules,
and Methods Using Chitosan Microcapsules in Cosmetic Preparations

BACKGROUND OF THE INVENTION--

Preliminary Amendment of U.S. National Stage for International Application PCT/EP00/07717 filed August 9, 2000

At page 2, line 11 thereof, please delete the section heading "Description of the Invention" and insert the following new section heading and new paragraph:

--BRIEF SUMMARY OF THE INVENTION

The present invention relates, in general, to decorative cosmetics and more particularly to preparations, more particularly makeup, lipsticks, mascaras, eye shadows and the like, which are distinguished by the fact that they contain microcapsules largely consisting of chitosan which are charged with special active substances.--

At page 2, line 24 thereof, please insert the following new section heading:

--DETAILED DESCRIPTION OF THE INVENTION--

At page 30, between lines 1 and 2, please add the following new paragraph:

--What is claimed is:--.

On a separate, new page 32, please add the following new section heading and paragraph containing an Abstract of the Disclosure:

--ABSTRACT OF THE DISCLOSURE

Cosmetic preparations, preferably decorative cosmetic preparations, comprising (a) at least one active substance present in the form of a chitosan microcapsule; and (b) one or more additives or auxiliaries are described. Methods of enhancing the stability and dermatological compatibility of such substances in cosmetic preparations by providing them in the form of chitosan microcapsules are also described.--

In the Claims:

Please add new claims 11-30, as follows:

--11. (New) A cosmetic preparation comprising:

(a) at least one active substance present in the form of a chitosan microcapsule; and

(b) one or more additives or auxiliaries.--

--12. (New) The cosmetic preparation according to claim 11, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, a chitosan and the at least one active substance; and contacting the matrix with an aqueous solution of an anionic polymer.--

--13. (New) The cosmetic preparation according to claim 11, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, a chitosan and the at least one active substance; dispersing the matrix in an oil phase; contacting the dispersed matrix with an aqueous solution of an anionic polymer; and removing the oil phase.--

--14. (New) The cosmetic preparation according to claim 11, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, an anionic polymer and the at least one active substance; and contacting the matrix with an aqueous solution of a chitosan.--

--15. (New) The cosmetic preparation according to claim 11, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, an anionic polymer and the at least one active substance; dispersing the matrix in an oil phase; contacting the dispersed matrix with an aqueous solution of a chitosan; and removing the oil phase.--

--16. (New) The cosmetic preparation according to claim 12, wherein the gel former comprises a component selected from the group consisting of heteropolysaccharides and proteins.--

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--17. (New) The cosmetic preparation according to claim 14, wherein the gel former comprises a component selected from the group consisting of heteropolysaccharides and proteins.--

--18. (New) The cosmetic preparation according to claim 12, wherein the anionic polymer comprises a component selected from the group consisting of alginic acid salts and anionic chitosan derivatives.--

--19. (New) The cosmetic preparation according to claim 14, wherein the gel former comprises a component selected from the group consisting of alginic acid salts and anionic chitosan derivatives.--

--20. (New) The cosmetic preparation according to claim 11, wherein the at least one active substance present in the form of a chitosan microcapsule is present in an amount of from 1 to 50% by weight based on the preparation.--

--21. (New) A method of enhancing the stability and dermatological compatibility of a decorative cosmetic preparation, said method comprising:

(a) providing an active substance in the form of a chitosan microcapsule; and

(b) combining the active substance with one or more additives or auxiliaries.--

--22. (New) The method according to claim 21, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, a chitosan and the at least one active substance; and contacting the matrix with an aqueous solution of an anionic polymer.--

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--23. (New) The method according to claim 21, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, a chitosan and the at least one active substance; dispersing the matrix in an oil phase; contacting the dispersed matrix with an aqueous solution of an anionic polymer; and removing the oil phase.--

--24. (New) The method according to claim 21, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, an anionic polymer and the at least one active substance; and contacting the matrix with an aqueous solution of a chitosan.--

--25. (New) The method according to claim 21, wherein the chitosan microcapsule is prepared by providing a matrix comprising a gel former, an anionic polymer and the at least one active substance; dispersing the matrix in an oil phase; contacting the dispersed matrix with an aqueous solution of a chitosan; and removing the oil phase.--

--26. (New) The method according to claim 22, wherein the gel former comprises a component selected from the group consisting of heteropolysaccharides and proteins.--

--27. (New) The method according to claim 24, wherein the gel former comprises a component selected from the group consisting of heteropolysaccharides and proteins.--

--28. (New) The method according to claim 22, wherein the anionic polymer comprises a component selected from the group consisting of alginic acid salts and anionic chitosan derivatives.--

Preliminary Amendment of U.S. National Stage for International Application PCT/EP00/07717 filed August 9, 2000

--29. (New) The method according to claim 24, wherein the gel former comprises a component selected from the group consisting of alginic acid salts and anionic chitosan derivatives.--

--30. (New) The method according to claim 21, wherein the at least one active substance present in the form of a chitosan microcapsule is present in an amount of from 1 to 50% by weight based on the preparation.--

Please cancel claims 1-10, without prejudice.

Preliminary Amendment of U.S. National Stage for International Application PCT/EP00/07717 filed August 9, 2000

REMARKS

Claims 11-30 are currently pending in the instant application.

The Specification has been amended to delete the original section headings and to insert the preferred section headings pursuant to 37 C.F.R. §1.77. A new Title of the Invention has been inserted. An Abstract of the Disclosure, in accordance with the disclosure, has been added. It is submitted that the amendments to the Specification made herein introduce no new matter. All of the amendments to the Specification constitute deletions of original section headings and/or paragraphs, and insertions or additions of new section headings and/or paragraphs. Accordingly, pursuant to 37 C.F.R. §1.121(b)(1)(iii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary. A separate page containing a clean copy of the Abstract of the Disclosure has been attached for the Examiner's convenience. Entry of the amendments to the Specification made herein are therefore proper and respectfully requested.

Original claims 1-10 have been canceled and replaced with new claims 11-30 solely for the purpose of improving clarity and grammar, which may suffer in translation, and not for any reason which relates to the statutory requirements for a patent. New claims 11-30 have not been added in response to any rejection, nor in anticipation of any rejection. Applicants respectfully submit that the scope of new claims 11-30 generally corresponds to the scope of original claims 1-10, and that new claims 11-30 are no narrower than original claims 1-10. Furthermore, although a moot point in view of their cancellation, Applicants respectfully submit that original claims 1-10 satisfied the requirements of 35 U.S.C. §112, as filed. New claims 11-30 are supported by the claims as originally filed and in the Specification, for example, at page 2, lines 12-23; at page 10, line 12, through page 12, line 19; and in the Examples. No new matter has been introduced. All of the amendments to the Claims constitute cancellation of original claims and the addition of new claims. Accordingly, pursuant to 37 C.F.R. §1.121(c)(1)(ii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary. Entry is therefore proper and respectfully requested.

Preliminary Amendment of U.S. National Stage for International Application PCT/EP00/07717 filed August 9, 2000

Prompt examination of the instant application in view of the amendments made herein is respectfully requested.

Respectfully submitted,

MARIA DeMORAGAS, *et al.*

February 19, 2002
(Date)

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ABSTRACT OF THE DISCLOSURE

Cosmetic preparations, preferably decorative cosmetic preparations, comprising (a) at least one active substance present in the form of a chitosan microcapsule; and (b) one or more additives or auxiliaries are described. Methods of enhancing the stability and dermatological compatibility of such substances in cosmetic preparations by providing them in the form of chitosan microcapsules are also described.

Decorative Cosmetic Preparations Containing Chitosan Microcapsules Charged with Active Ingredients

Field of the Invention

This invention relates generally to decorative cosmetics and more particularly to preparations, more particularly makeup, lipsticks, mascaras, eye shadows and the like, which are distinguished by the fact that they contain microcapsules largely consisting of chitosan which are charged with special active substances.

Prior Art

The desire to appear beautiful and more attractive has been rooted in mankind for thousands of years. Although the preparations with which this is achieved have constantly changed, even modern decorative personal care preparations contain a more or less large percentage of dyes which change the color of the face, the eye region, the lips and the nails. In addition, special ingredients perform additional skin-care and skin-protecting functions. The dyes used include white pigments, such as talcum, zinc oxide, kaolin, titanium dioxide, inorganic pigments, such as iron oxides, chromium oxides, ultramarine, manganese violet, and organic colored pigments. In addition, such pigments as bismuth oxychloride, mica, titanium-dioxide-coated mica and pearl essence, which produce a pearlescent effect, are frequently used. Under the law, dyes used in the eye and lip region must have the appropriate mucous membrane compatibility. The wide variety of dyes which is significantly increased by the number of care substances makes the formulation of decorative cosmetic products expensive and problematical. The substances often cannot be homogeneously incorporated so that separation occurs - particularly in the event of prolonged storage or in heat - and, although not

immediately spoiling the product, makes it unattractive. It will readily be appreciated that a consumer who buys a lipstick that becomes patchy in heat or a nail varnish that separates after a few weeks would not repeat the purchase.

Accordingly, the problem addressed by the present invention was to provide decorative cosmetic preparations containing active substances, more particularly pigments, in an easy-to-formulate heat-, storage- and surfactant-stable form which would show high dermatological compatibility and, at the same time, would have a particularly attractive appearance.

Description of the Invention

The present invention relates to decorative cosmetic preparations containing chitosan microcapsules charged with active substances.

It has surprisingly been found that the encapsulation of active substances - both colored pigments and care components - eminently satisfies the stated requirements. The capsules are stable to heat, storage and surfactants, are easy to formulate and only release the active substance under mechanical pressure, i.e. on application to the skin or nails. An additional care effect and high cosmetic compatibility, particularly in the region of the sensitive ophthalmic mucous membrane, are achieved by the use of chitosan as the key constituent of the capsules. In addition, the microcapsules charged with various dyes provide the preparations with an interesting appearance.

Active substances

Basically, the choice of the active substances encapsulated in the new microcapsules is not critical. They are preferably substances which are only released by mechanical destruction of the microcapsules. It may be that the encapsulated substances are not to be released at all and merely serve the purpose of providing the preparation with an aesthetic

appearance. This often applies, for example, to dyes. Typical examples of active substances used in decorative cosmetic preparations are cosmetic oils, pearlizing waxes, stabilizers, biogenic agents, vitamins, UV protection factors, antioxidants, preservatives, insect repellents, self-tanning agents, tyrosine inhibitors (depigmenting agents), perfume oils and dyes.

Suitable **cosmetic oils** are, for example, Guerbet alcohols based on fatty alcohols containing 6 to 18 and preferably 8 to 10 carbon atoms, esters of linear C_{6-22} fatty acids with linear C_{6-22} fatty alcohols, esters of branched C_{6-13} carboxylic acids with linear C_{6-22} fatty alcohols such as, for example, myristyl myristate, myristyl palmitate, myristyl stearate, myristyl isostearate, myristyl oleate, myristyl behenate, myristyl erucate, cetyl myristate, cetyl palmitate, cetyl stearate, cetyl isostearate, cetyl oleate, cetyl behenate, cetyl erucate, stearyl myristate, stearyl palmitate, stearyl stearate, stearyl isostearate, stearyl oleate, stearyl behenate, stearyl erucate, isostearyl myristate, isostearyl palmitate, isostearyl stearate, isostearyl isostearate, isostearyl oleate, isostearyl behenate, isostearyl oleate, oleyl myristate, oleyl palmitate, oleyl stearate, oleyl isostearate, oleyl oleate, oleyl behenate, oleyl erucate, behenyl myristate, behenyl palmitate, behenyl stearate, behenyl isostearate, behenyl oleate, behenyl behenate, behenyl erucate, erucyl myristate, erucyl palmitate, erucyl stearate, erucyl isostearate, erucyl oleate, erucyl behenate and erucyl erucate. Also suitable are esters of linear C_{6-22} fatty acids with branched alcohols, more particularly 2-ethyl hexanol, esters of hydroxycarboxylic acids with linear or branched C_{6-22} fatty alcohols, more especially Dioctyl Malate, esters of linear and/or branched fatty acids with polyhydric alcohols (for example propylene glycol, dimer diol or trimer triol) and/or Guerbet alcohols, triglycerides based on C_{6-10} fatty acids, liquid mono-/di-/tri-glyceride mixtures based on C_{6-18} fatty acids, esters of C_{6-22} fatty alcohols and/or Guerbet alcohols with aromatic carboxylic acids, more particularly benzoic acid, esters of C_{2-12} dicarboxylic acids with linear or branched

alcohols containing 1 to 22 carbon atoms or polyols containing 2 to 10 carbon atoms and 2 to 6 hydroxyl groups, vegetable oils, branched primary alcohols, substituted cyclohexanes, linear and branched C_{6-22} fatty alcohol carbonates, Guerbet carbonates, esters of benzoic acid with linear and/or branched C_{6-22} alcohols (for example Finsolv® TN), linear or branched, symmetrical or nonsymmetrical dialkyl ethers containing 6 to 22 carbon atoms per alkyl group, ring opening products of epoxidized fatty acid esters with polyols, silicone oils and/or aliphatic or naphthenic hydrocarbons, for example squalane, squalene or dialkyl cyclohexanes.

Suitable **pearlizing waxes** are, for example, alkylene glycol esters, especially ethylene glycol distearate; fatty acid alkanolamides, especially cocofatty acid diethanolamide; partial glycerides, especially stearic acid monoglyceride; esters of polybasic, optionally hydroxysubstituted carboxylic acids with fatty alcohols containing 6 to 22 carbon atoms, especially long-chain esters of tartaric acid; fatty compounds, such as for example fatty alcohols, fatty ketones, fatty aldehydes, fatty ethers and fatty carbonates which contain in all at least 24 carbon atoms, especially laurone and distearylether; fatty acids, such as stearic acid, hydroxystearic acid or behenic acid, ring opening products of olefin epoxides containing 12 to 22 carbon atoms with fatty alcohols containing 12 to 22 carbon atoms and/or polyols containing 2 to 15 carbon atoms and 2 to 10 hydroxyl groups and mixtures thereof.

Metal salts of fatty acids such as, for example, magnesium, aluminium and/or zinc stearate or ricinoleate may be used as **stabilizers**.

In the context of the invention, **biogenic agents** are, for example, tocopherol, tocopherol acetate, tocopherol palmitate, ascorbic acid, deoxyribonucleic acid, retinol, bisabolol, allantoin, phytantriol, panthenol, AHA acids, amino acids, ceramides, pseudoceramides, essential oils, plant extracts and vitamin complexes.

Examples of **UV protection factors are** organic substances (light

filters) which are liquid or crystalline at room temperature and which are capable of absorbing ultraviolet radiation and of releasing the energy absorbed in the form of longer-wave radiation, for example heat. UV-B filters can be oil-soluble or water-soluble. The following are examples of oil-soluble substances:

- 3-benzylidene camphor or 3-benzylidene norcamphor and derivatives thereof, for example 3-(4-methylbenzylidene)-camphor, as described in **EP 0693471 B1**;
- 4-aminobenzoic acid derivatives, preferably 4-(dimethylamino)-benzoic acid-2-ethylhexyl ester, 4-(dimethylamino)-benzoic acid-2-octyl ester and 4-(dimethylamino)-benzoic acid amyl ester;
- esters of cinnamic acid, preferably 4-methoxycinnamic acid-2-ethylhexyl ester, 4-methoxycinnamic acid propyl ester, 4-methoxycinnamic acid isoamyl ester, 2-cyano-3,3-phenylcinnamic acid-2-ethylhexyl ester (Octocrylene);
- esters of salicylic acid, preferably salicylic acid-2-ethylhexyl ester, salicylic acid-4-isopropylbenzyl ester, salicylic acid homomenthyl ester;
- derivatives of benzophenone, preferably 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-methoxy-4'-methylbenzophenone, 2,2'-dihydroxy-4-methoxybenzophenone;
- esters of benzalmalonic acid, preferably 4-methoxybenzalmalonic acid di-2-ethylhexyl ester;
- triazine derivatives such as, for example, 2,4,6-trianilino-(p-carbo-2'-ethyl-1'-hexyloxy)-1,3,5-triazine and Octyl Triazone, as described in **EP 0 818 450 A1**, or Dioctyl Butamido Triazine (Uvasorb® HEB);
- propane-1,3-diones such as, for example, 1-(4-tert.butylphenyl)-3-(4'-methoxyphenyl)-propane-1,3-dione;
- ketotricyclo(5.2.1)decane derivatives, as described in **EP 0 694 521 B1**.

Suitable water-soluble substances are

- 2-phenylbenzimidazole-5-sulfonic acid and alkali metal, alkaline earth metal, ammonium, alkylammonium, alkanolammonium and glucammonium salts thereof;
- sulfonic acid derivatives of benzophenones, preferably 2-hydroxy-4-methoxybenzophenone-5-sulfonic acid and salts thereof;
- sulfonic acid derivatives of 3-benzylidene camphor such as, for example, 4-(2-oxo-3-bornylidenemethyl)-benzene sulfonic acid and 2-methyl-5-(2-oxo-3-bornylidene)-sulfonic acid and salts thereof.

Typical UV-A filters are, in particular, derivatives of benzoyl methane such as, for example 1-(4'-tert.butylphenyl)-3-(4'-methoxyphenyl)-propane-1,3-dione, 4-tert-butyl-4'-methoxydibenzoylmethane (Parsol 1789), 1-phenyl-3-(4'-isopropylphenyl)-propane-1,3-dione and the eneamine compounds described in **DE 19712033 A1** (BASF). The UV-A and UV-B filters may of course also be used in the form of mixtures. Besides the soluble substances mentioned, insoluble pigments, i.e. finely dispersed metal oxides or salts, may also be used for this purpose. Examples of suitable metal oxides are, in particular, zinc oxide and titanium dioxide and also oxides of iron, zirconium, silicon, manganese, aluminium and cerium and mixtures thereof. Silicates (talcum), barium sulfate and zinc stearate may be used as salts. The oxides and salts are used in the form of the pigments for skin-care and skin-protecting emulsions and decorative cosmetics. The particles should have an average diameter of less than 100 nm, preferably from 5 to 50 nm and more preferably from 15 to 30 nm. They may be spherical in shape although ellipsoidal particles or other non-spherical particles may also be used. The pigments may also be surface-treated, i.e. hydrophilicized or hydrophobicized. Typical examples are coated titanium dioxides such as, for example, Titandioxid T 805 (Degussa)

or Eusolex® T2000 (Merck). Suitable hydrophobic coating materials are, above all, silicones and particularly trialkoxyoctyl silanes or simethicones. So-called micro- or nanopigments are preferably used in sun protection products. Micronized zinc oxide is preferably used. Other suitable UV filters can be found in P. Finkel's review in **SÖFW-Journal 122, 543 (1996)**.

Besides the two above-mentioned groups of primary protection factors, secondary protection factors of the **antioxidant** type may also be used. Secondary sun protection factors of the antioxidant type interrupt the photochemical reaction chain which is initiated when UV rays penetrate into the skin. Typical examples of suitable antioxidants are amino acids (for example glycine, histidine, tyrosine, tryptophane) and derivatives thereof, imidazoles (for example urocanic acid) and derivatives thereof, peptides, such as D,L-carnosine, D-carnosine, L-carnosine and derivatives thereof (for example anserine), carotinoids, carotenes (for example α-carotene, β-carotene, lycopene) and derivatives thereof, chlorogenic acid and derivatives thereof, liponic acid and derivatives thereof (for example dihydroliponic acid), aurothioglucose, propylthiouracil and other thiols (for example thioredoxine, glutathione, cysteine, cystine, cystamine and glycosyl, N-acetyl, methyl, ethyl, propyl, amyl, butyl and lauryl, palmitoyl, oleyl, γ-linoleyl, cholesteryl and glyceryl esters thereof) and their salts, dilaurylthiodipropionate, distearylthiodipropionate, thiodipropionic acid and derivatives thereof (esters, ethers, peptides, lipids, nucleotides, nucleosides and salts) and sulfoximine compounds (for example butionine sulfoximines, homocysteine sulfoximine, butionine sulfones, penta-, hexa- and hepta-thionine sulfoximine) in very small compatible dosages (for example pmole to μmole/kg), also (metal) chelators (for example α-hydroxyfatty acids, palmitic acid, phytic acid, lactoferrine), α-hydroxy acids (for example citric acid, lactic acid, malic acid), humic acid, bile acid, bile extracts, bilirubin, biliverdin, EDTA, EGTA and derivatives thereof,

unsaturated fatty acids and derivatives thereof (for example γ-linolenic acid, linoleic acid, oleic acid), folic acid and derivatives thereof, ubiquinone and ubiquinol and derivatives thereof, vitamin C and derivatives thereof (for example ascorbyl palmitate, Mg ascorbyl phosphate, ascorbyl acetate), liponic acid, tocopherols and derivatives (for example vitamin E acetate), vitamin A and derivatives (vitamin A palmitate) and coniferyl benzoate of benzoin resin, rutinic acid and derivatives thereof, α-glycosyl rutin, ferulic acid, furfurylidene glucitol, carnosine, butyl hydroxytoluene, butyl hydroxyanisole, nordihydroguaiac resin acid, nordihydroguaiaretic acid, trihydroxybutyrophenone, uric acid and derivatives thereof, mannose and derivatives thereof, Superoxid-Dismutase, zinc and derivatives thereof (for example ZnO, $ZnSO_4$), selenium and derivatives thereof (for example selenium methionine), stilbenes and derivatives thereof (for example stilbene oxide, trans-stilbene oxide) and derivatives of these active substances suitable for the purposes of the invention (salts, esters, ethers, sugars, nucleotides, nucleosides, peptides and lipids).

Suitable **preservatives** are, for example, phenoxyethanol, formaldehyde solution, parabens, pentanediol or sorbic acid and the other classes of compounds listed in Appendix 6, Parts A and B of the Kosmetikverordnung ("Cosmetics Directive"). Suitable **insect repellents** are N,N-diethyl-m-toluamide, pentane-1,2-diol or Ethyl Butylacetyl-aminopropionate. A suitable **self-tanning agent** is dihydroxyacetone. **Tyrosine inhibitors**, which prevent the formation of melanin and are used in depigmenting formulations, are for example arbutin, koji acid, coumaric acid and ascorbic acid (vitamin C)

Suitable **perfume oils** are mixtures of natural and synthetic perfumes. Natural perfumes include the extracts of blossoms (lily, lavender, rose, jasmine, neroli, ylang-ylang), stems and leaves (geranium, patchouli, petitgrain), fruits (anise, coriander, caraway, juniper), fruit peel (bergamot, lemon, orange), roots (nutmeg, angelica, celery, cardamon,

costus, iris, calmus), woods (pinewood, sandalwood, guaiac wood, cedarwood, rosewood), herbs and grasses (tarragon, lemon grass, sage, thyme), needles and branches (spruce, fir, pine, dwarf pine), resins and balsams (galbanum, elemi, benzoin, myrrh, olibanum, opoponax). Animal raw materials, for example civet and beaver, may also be used. Typical synthetic perfume compounds are products of the ester, ether, aldehyde, ketone, alcohol and hydrocarbon type. Examples of perfume compounds of the ester type are benzyl acetate, phenoxyethyl isobutyrate, p-tert.butyl cyclohexylacetate, linalyl acetate, dimethyl benzyl carbinyl acetate, phenyl ethyl acetate, linalyl benzoate, benzyl formate, ethylmethyl phenyl glycinate, allyl cyclohexyl propionate, styrallyl propionate and benzyl salicylate. Ethers include, for example, benzyl ethyl ether while aldehydes include, for example, the linear alkanals containing 8 to 18 carbon atoms, citral, citronellal, citronellyloxyacetaldehyde, cyclamen aldehyde, hydroxycitronellal, lilial and bourgeonal. Examples of suitable ketones are the ionones, α-isomethylionone and methyl cedryl ketone. Suitable alcohols are anethol, citronellol, eugenol, isoeugenol, geraniol, linalool, phenylethyl alcohol and terpineol. The hydrocarbons mainly include the terpenes and balsams. However, it is preferred to use mixtures of different perfume compounds which, together, produce an agreeable fragrance. Other suitable perfume oils are essential oils of relatively low volatility which are mostly used as aroma components. Examples are sage oil, camomile oil, clove oil, melissa oil, mint oil, cinnamon leaf oil, lime-blossom oil, juniper berry oil, vetiver oil, olibanum oil, galbanum oil, ladanum oil and lavendin oil. The following are preferably used either individually or in the form of mixtures: bergamot oil, dihydromyrcenol, lilial, lyral, citronellol, phenylethyl alcohol, α-hexylcinnamaldehyde, geraniol, benzyl acetone, cyclamen aldehyde, linalool, Boisambrene Forte, Ambroxan, indole, hedione, sandelice, citrus oil, mandarin oil, orange oil, allylamyl glycolate, cyclovertal, lavendin oil, clary oil, β-damascone, geranium oil bourbon,

cyclohexyl salicylate, Vertofix Coeur, Iso-E-Super, Fixolide NP, evernyl, iraldein gamma, phenylacetic acid, geranyl acetate, benzyl acetate, rose oxide, romillat, irotyl and floramat.

Suitable **dyes** are any of the substances suitable and approved for cosmetic purposes as listed, for example, in the publication **"Kosmetische Färbemittel"** of the **Farbstoffkommission der Deutschen Forschungsgemeinschaft, Verlag Chemie, Weinheim, 1984, pages 81 to 106.** These active substances may also be present in the capsules solely for aesthetic reasons, i.e are not intended for controlled release.

Chitosan microcapsules

"Microcapsules" are understood to be aggregates with a diameter of about 0.1 to about 5 mm which contain at least one solid or liquid core surrounded by at least one continuous membrane. More precisely, they are finely dispersed liquid or solid phases coated with film-forming polymers, in the production of which the polymers are deposited onto the material to be encapsulated after emulsification and coacervation or interfacial polymerization. In another process, liquid active substances are absorbed in a matrix ("microsponge") which, as microparticles, may be additionally coated with film-forming polymers. The microscopically small capsules, also known as nanocapsules, can be dried in the same way as powders. Besides single-core microcapsules, there are also multiple-core aggregates, also known as microspheres, which contain two or more cores distributed in the continuous membrane material. In addition, single-core or multiple-core microcapsules may be surrounded by an additional second, third etc. membrane. The membrane may consist of natural, semisynthetic or synthetic materials. Natural membrane materials are, for example, gum arabic, agar agar, agarose, maltodextrins, alginic acid and salts thereof, for example sodium or calcium alginate, fats and fatty acids, cetyl alcohol, collagen, chitosan, lecithins, gelatin, albumin, shellac, polysaccharides,

such as starch or dextran, sucrose and waxes. Semisynthetic membrane materials are inter alia chemically modified celluloses, more particularly cellulose esters and ethers, for example cellulose acetate, ethyl cellulose, hydroxypropyl cellulose, hydroxypropyl methyl cellulose and carboxymethyl cellulose, and starch derivatives, more particularly starch ethers and esters. Synthetic membrane materials are, for example, polymers, such as polyacrylates, polyamides, polyvinyl alcohol or polyvinyl pyrrolidone.

Examples of known microcapsules are the following commercial products (the membrane material is shown in brackets) *Hallcrest Microcapsules* (gelatin, gum arabic), *Coletica Thalaspheres* (maritime collagen), *Lipotec Millicapseln* (alginic acid, agar agar), *Induchem Unispheres* (lactose, microcrystalline cellulose, hydroxypropylmethyl cellulose), *Unicerin C30* (lactose, microcrystalline cellulose, hydroxypropylmethyl cellulose), *Kobo Glycospheres* (modified starch, fatty acid esters, phospholipids), *Softspheres* (modified agar agar) and *Kuhs Probiol Nanospheres* (phospholipids).

Reference is also made in this connection to German patent application **DE 19712978 A1** (Henkel) which describes chitosan microspheres obtained by mixing chitosans or chitosan derivatives with oil components and introducing the resulting mixtures into alkalized surfactant solutions. In addition, the use of chitosan as an encapsulating material for tocopherol is known from German patent application **DE 19756452 A1** (Henkel). Chitosan microcapsules and processes for their production are the subject of earlier patent applications filed by applicants. There are essentially two processes for this purpose:

(1) microcapsules with mean diameters of 0.1 to 5 mm consisting of a membrane and a matrix containing at least one active substance and obtainable by

(a) preparing a matrix from gel formers, chitosans and active substances,
(b) optionally dispersing the matrix in an oil phase and
(c) treating the dispersed matrix with aqueous solutions of anionic polymers and optionally removing the oil phase in the process;

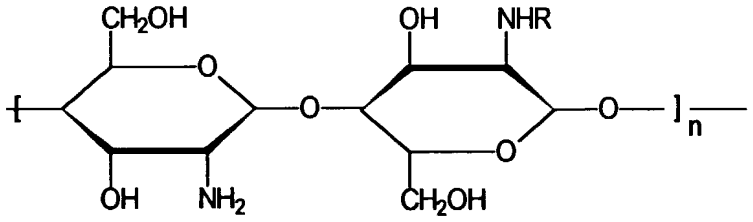
(2) microcapsules with mean diameters of 0.1 to 5 mm consisting of a membrane and a matrix containing at least one active substance and obtainable by

(a) preparing a matrix from gel formers, anionic polymers and active substances,
(b) optionally dispersing the matrix in an oil phase and
(c) treating the dispersed matrix with aqueous chitosan solutions and optionally removing the oil phase in the process;

Preferred **gel formers** are substances which are capable of forming gels in aqueous solution at temperatures above 40°C. Typical examples of such gel formers are heteropolysaccharides and proteins. Preferred thermogelling heteropolysaccharides are agaroses which may be present in the form of the agar agar obtainable from red algae, even together with up to 30% by weight of non-gel-forming agaropectins. The principal constituent of agaroses are linear polysaccharides of D-galactose and 3,6-anhydro-L-galactose with alternate β-1,3- and β-1,4-glycosidic bonds. The heteropolysaccharides preferably have a molecular weight of 110,000 to 160,000 and are both odorless and tasteless. Suitable alternatives are pectins, xanthans (including xanthan gum) and mixtures thereof. Other preferred types are those which - in 1% by weight aqueous solution - still form gels that do not melt below 80°C and solidify again above 40°C. Examples from the group of thermogelling proteins are the various

gelatines.

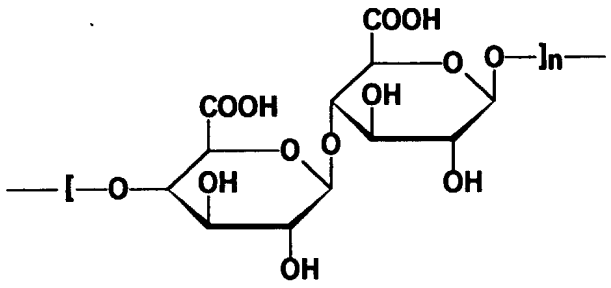
Chitosans are biopolymers which belong to the group of hydrocolloids. Chemically, they are partly deacetylated chitins differing in their molecular weights which contain the following – idealized – monomer unit:



In contrast to most hydrocolloids, which are negatively charged at biological pH values, chitosans are cationic biopolymers under these conditions. The positively charged chitosans are capable of interacting with oppositely charged surfaces and are therefore used in cosmetic hair-care and body-care products and pharmaceutical preparations (cf. **Ullmann's Encyclopedia of Industrial Chemistry,** 5th Ed., Vol. A6, Weinheim, Verlag Chemie, 1986, pages 231-332). Overviews of this subject have also been published, for example, by B. Gesslein et al. in **HAPPI** 27, 57 (1990), O. Skaugrud in **Drug Cosm. Ind.** 148, 24 (1991) and E. Onsoyen et al. in **Seifen-Öle-Fette-Wachse** 117, 633 (1991). Chitosans are produced from chitin, preferably from the shell residues of crustaceans which are available in large quantities as inexpensive raw materials. In a process described for the first time by Hackmann et al., the chitin is normally first deproteinized by addition of bases, demineralized by addition of mineral acids and, finally, deacetylated by addition of strong bases, the molecular weights being distributed over a broad spectrum. Corresponding processes are known, for example, from **Makromol. Chem.** 177, 3589 (1976) or French patent application **FR 2701266 A**. Preferred types are those which are disclosed in German patent applications **DE 4442987 A1** and **DE 19537001 A1**

(Henkel) and which have an average molecular weight of 10,000 to 500,000 dalton or 800,000 to 1,200,000 dalton and/or a Brookfield viscosity (1% by weight in glycolic acid) below 5,000 mPas, a degree of deacetylation of 80 to 88% and an ash content of less than 0.3% by weight. In the interests of better solubility in water, the chitosans are generally used in the form of their salts, preferably as glycolates.

The function of the **anionic polymers** is to form membranes with the chitosans. Depending on the production process, they may be present in the matrix (in which case the membrane is formed by treatment with the chitosan solutions) or may serve as precipitant for the chitosans present in the matrix. Preferred anionic polymers are salts of alginic acid. The alginic acid is a mixture of carboxyl-containing polysaccharides with the following idealized monomer unit:



The average molecular weight of the alginic acid or the alginates is in the range from 150,000 to 250,000. Salts of alginic acid and complete and partial neutralization products thereof are understood in particular to be the alkali metal salts, preferably sodium alginate ("algin") and the ammonium and alkaline earth metal salts. Mixed alginates, for example sodium/magnesium or sodium/calcium alginates, are particularly preferred. In an alternative embodiment of the invention, however, anionic chitosan derivatives, for example the carboxylation and above all succinylation products described, for example, in German patent **DE 3713099 C2**

(L'Oréal) and German patent application **DE 19604180 A1** (Henkel) are also suitable for this purpose.

To produce the chitosan microcapsules, a 1 to 10 and preferably 2 to 5% by weight aqueous solution of the gel former, preferably agar agar, is normally prepared and heated under reflux. A second aqueous solution containing the anionic polymers in quantities of 0.1 to 2 and preferably 0.25 to 0.5% by weight and the active substance in quantities of 0.1 to 25 and preferably 0.25 to 10% by weight is added in the boiling heat, preferably at 80 to 100°C; this mixture is called the matrix. Accordingly, the charging of the microcapsules with active substances may also comprise 0.1 to 25% by weight, based on the weight of the capsules. If desired, water-insoluble constituents, for example inorganic pigments, may be added at this stage to adjust viscosity, generally in the form of aqueous or aqueous/alcoholic dispersions. In addition, to emulsify or disperse the active substances, it can be useful to add emulsifiers and/or solubilizers to the matrix.

Suitable **emulsifiers** are, for example, nonionic surfactants from at least one of the following groups:

- products of the addition of 2 to 30 moles of ethylene oxide and/or 0 to 5 moles of propylene oxide onto linear C_{8-22} fatty alcohols, C_{12-22} fatty acids and alkyl phenols containing 8 to 15 carbon atoms in the alkyl group and alkylamines containing 8 to 22 carbon atoms in the alkyl group;
- alkyl and/or alkenyl oligoglycosides containing 8 to 22 carbon atoms in the alkyl group and ethoxylated analogs thereof;
- addition products of 1 to 15 moles of ethylene oxide with castor oil and/or hydrogenated castor oil;
- addition products of 15 to 60 moles of ethylene oxide with castor oil and/or hydrogenated castor oil;
- partial esters of glycerol and/or sorbitan with unsaturated, linear or

saturated, branched fatty acids containing 12 to 22 carbon atoms and/or hydroxycarboxylic acids containing 3 to 18 carbon atoms and addition products thereof with 1 to 30 moles of ethylene oxide;

- partial esters of polyglycerol (average degree of self-condensation 2 to 8), polyethylene glycol (molecular weight 400 to 5000), trimethylolpropane, pentaerythritol, sugar alcohols (for example sorbitol), alkyl glucosides (for example methyl glucoside, butyl glucoside, lauryl glucoside) and polyglucosides (for example cellulose) with saturated and/or unsaturated, linear or branched fatty acids containing 12 to 22 carbon atoms and/or hydroxycarboxylic acids containing 3 to 18 carbon atoms and addition products thereof with 1 to 30 moles of ethylene oxide;
- mixed esters of pentaerythritol, fatty acids, citric acid and fatty alcohol according to **DE-PS 11 65 574** and/or mixed esters of fatty acids containing 6 to 22 carbon atoms, methyl glucose and polyols, preferably glycerol or polyglycerol,
- mono-, di- and trialkyl phosphates and mono-, di- and/or tri-PEG-alkyl phosphates and salts thereof,
- wool wax alcohols,
- polysiloxane/polyalkyl/polyether copolymers and corresponding derivatives,
- polyalkylene glycols and
- glycerol carbonate.

The **addition products of ethylene oxide and/or propylene oxide** with fatty alcohols, fatty acids, alkylphenols or with castor oil are known commercially available products. They are homolog mixtures of which the average degree of alkoxylation corresponds to the ratio between the quantities of ethylene oxide and/or propylene oxide and substrate with which the addition reaction is carried out. $C_{12/18}$ fatty acid monoesters and

diesters of addition products of ethylene oxide with glycerol are known as refatting agents for cosmetic formulations from **DE-PS 20 24 051**.

Alkyl and/or alkenyl oligoglycosides, their production and their use are known from the prior art. They are produced in particular by reacting glucose or oligosaccharides with primary alcohols containing 8 to 18 carbon atoms. So far as the glucoside unit is concerned, both monoglycosides in which a cyclic sugar unit is attached to the fatty alcohol by a glycoside bond and oligomeric glycosides with a degree of oligomerization of preferably up to about 8 are suitable. The degree of oligomerization is a statistical mean value on which the homolog distribution typical of such technical products is based.

Typical examples of suitable **partial glycerides** are hydroxystearic acid monoglyceride, hydroxystearic acid diglyceride, isostearic acid monoglyceride, isostearic acid diglyceride, oleic acid monoglyceride, oleic acid diglyceride, ricinoleic acid monoglyceride, ricinoleic acid diglyceride, linoleic acid monoglyceride, linoleic acid diglyceride, linolenic acid monoglyceride, linolenic acid diglyceride, erucic acid monoglyceride, erucic acid diglyceride, tartaric acid monoglyceride, tartaric acid diglyceride, citric acid monoglyceride, citric acid diglyceride, malic acid monoglyceride, malic acid diglyceride and technical mixtures thereof which may still contain small quantities of triglyceride from the production process. Addition products of 1 to 30 and preferably 5 to 10 moles of ethylene oxide with the partial glycerides mentioned are also suitable.

Suitable **sorbitan esters** are sorbitan monoisostearate, sorbitan sesquiisostearate, sorbitan diisostearate, sorbitan triisostearate, sorbitan monooleate, sorbitan sesquioleate, sorbitan dioleate, sorbitan trioleate, sorbitan monoerucate, sorbitan sesquierucate, sorbitan dierucate, sorbitan trierucate, sorbitan monoricinoleate, sorbitan sesquiricinoleate, sorbitan diricinoleate, sorbitan triricinoleate, sorbitan monohydroxystearate, sorbitan sesquihydroxystearate, sorbitan dihydroxystearate, sorbitan trihydroxy-

stearate, sorbitan monotartrate, sorbitan sesquitartrate, sorbitan ditartrate, sorbitan tritartrate, sorbitan monocitrate, sorbitan sesquicitrate, sorbitan dicitrate, sorbitan tricitrate, sorbitan monomaleate, sorbitan sesquimaleate, sorbitan dimaleate, sorbitan trimaleate and technical mixtures thereof. Addition products of 1 to 30 and preferably 5 to 10 moles of ethylene oxide with the sorbitan esters mentioned are also suitable.

Typical examples of suitable **polyglycerol esters** are Polyglyceryl-2 Dipolyhydroxystearate (Dehymuls® PGPH), Polyglycerin-3-Diisostearate (Lameform® TGI), Polyglyceryl-4 Isostearate (Isolan® GI 34), Polyglyceryl-3 Oleate, Diisostearoyl Polyglyceryl-3 Diisostearate (Isolan® PDI), Polyglyceryl-3 Methylglucose Distearate (Tego Care® 450), Polyglyceryl-3 Beeswax (Cera Bellina®), Polyglyceryl-4 Caprate (Polyglycerol Caprate T2010/90), Polyglyceryl-3 Cetyl Ether (Chimexane® NL), Polyglyceryl-3 Distearate (Cremophor® GS 32) and Polyglyceryl Polyricinoleate (Admul® WOL 1403), Polyglyceryl Dimerate Isostearate and mixtures thereof.

Examples of other suitable **polyolesters** are the mono-, di- and triesters of trimethylol propane or pentaerythritol with lauric acid, cocofatty acid, tallow fatty acid, palmitic acid, stearic acid, oleic acid, behenic acid and the like optionally reacted with 1 to 30 moles of ethylene oxide.

Other suitable emulsifiers are **zwitterionic surfactants**. Zwitterionic surfactants are surface-active compounds which contain at least one quaternary ammonium group and at least one carboxylate and one sulfonate group in the molecule. Particularly suitable zwitterionic surfactants are the so-called betaines, such as the N-alkyl-N,N-dimethyl ammonium glycinates, for example cocoalkyl dimethyl ammonium glycinate, N-acylaminopropyl-N,N-dimethyl ammonium glycinates, for example cocoacylaminopropyl dimethyl ammonium glycinate, and 2-alkyl-3-carboxymethyl-3-hydroxyethyl imidazolines containing 8 to 18 carbon atoms in the alkyl or acyl group and cocoacylaminoethyl hydroxyethyl carboxymethyl glycinate. The fatty acid amide derivative known under the

CTFA name of *Cocamidopropyl Betaine* is particularly preferred. Ampholytic surfactants are also suitable emulsifiers. Ampholytic surfactants are surface-active compounds which, in addition to a $C_{8/18}$ alkyl or acyl group, contain at least one free amino group and at least one -COOH- or -SO_3H- group in the molecule and which are capable of forming inner salts. Examples of suitable ampholytic surfactants are N-alkyl glycines, N-alkyl propionic acids, N-alkylaminobutyric acids, N-alkyliminodipropionic acids, N-hydroxyethyl-N-alkylamidopropyl glycines, N-alkyl taurines, N-alkyl sarcosines, 2-alkylaminopropionic acids and alkylaminoacetic acids containing around 8 to 18 carbon atoms in the alkyl group. Particularly preferred ampholytic surfactants are N-cocoalkylaminopropionate, cocoacylaminoethyl aminopropionate and $C_{12/18}$ acyl sarcosine.

Finally, other suitable emulsifiers are **cationic surfactants**, those of the esterquat type, preferably methyl-quaternized difatty acid triethanolamine ester salts, being particularly preferred.

Suitable solubilizers or **hydrotropes** are, for example, ethanol, isopropyl alcohol or polyols. Suitable polyols preferably contain 2 to 15 carbon atoms and at least two hydroxyl groups. The polyols may contain other functional groups, more especially amino groups, or may be modified with nitrogen. Typical examples are

- glycerol;
- alkylene glycols such as, for example, ethylene glycol, diethylene glycol, propylene glycol, butylene glycol, hexylene glycol and polyethylene glycols with an average molecular weight of 100 to 1000 dalton;
- technical oligoglycerol mixtures with a degree of self-condensation of 1.5 to 10 such as, for example, technical diglycerol mixtures with a diglycerol content of 40 to 50% by weight;
- methylol compounds such as, in particular, trimethylol ethane, trimethylol propane, trimethylol butane, pentaerythritol and

dipentaerythritol;

- lower alkyl glucosides, particularly those containing 1 to 8 carbon atoms in the alkyl group, for example methyl and butyl glucoside;
- sugar alcohols containing 5 to 12 carbon atoms, for example sorbitol or mannitol,
- sugars containing 5 to 12 carbon atoms, for example glucose or sucrose;
- amino sugars, for example glucamine;
- dialcoholamines, such as diethanolamine or 2-aminopropane-1,3-diol.

The concentration of emulsifiers may be in the range from 1 to 20% by weight and is preferably in the range from 5 to 10% by weight, based on the active substances. The quantity of solubilizers is determined solely by the solubility or dispersibility of the active substances in water.

After its preparation from gel former, chitosan and active substance, the matrix is very finely dispersed in an oil phase with intensive shearing in order to produce small particles in the subsequent encapsulation process. It has proved to be particularly advantageous in this regard to heat the matrix to temperatures in the range from 40 to 60°C while the oil phase is cooled to 10 to 20°C. The actual encapsulation, i.e. formation of the membrane by contacting the chitosan in the matrix with the anionic polymers, takes place in the third step. To this end, it is advisable to wash the matrix dispersed in the oil phase with an aqueous ca. 0.1 to 3 and preferably 0.25 to 0.5% by weight aqueous solution of the anionic polymer, preferably the alginate, at a temperature in the range from 40 to 100 and preferably 50 to 60°C and, at the same time, to remove the oil phase.

Similarly, a matrix of the gel former, anionic polymer and active substance can be formed in the first step and dispersed in an oil phase and the capsules subsequently prepared by precipitation with a chitosan solution. To this end, it is sufficient merely to interchange the "anionic

polymer" and the "chitosan" in the above-described production process and to keep to the same quantities. In another two alternative embodiments, dispersion in an oil phase can be dispensed with although this does result in larger capsules. Accordingly, there are in all four processes for producing the chitosan microcapsules. The resulting aqueous preparations generally have a microcapsule content of 1 to 10% by weight. In some cases, it can be of advantage for the solution of the polymers to contain other substances, for example emulsifiers or preservatives. After filtration, microcapsules with a mean diameter of preferably 1 to 3 mm are obtained. It is advisable to sieve the capsules to ensure a uniform size distribution. The microcapsules thus obtained may have any shape within production-related limits, but are preferably substantially spherical.

Decorative cosmetic preparations

Chitosan microcapsules charged with active substances have been found to be particularly stable in storage and stable to surfactants and, at the same time, to show high dermatological compatibility. The active substances are stably encapsulated and are only released under mechanical pressure. The chitosan microcapsules charged with active substances are thus particularly suitable for the production of decorative cosmetic preparations such as, for example, makeup, rouges, lipsticks, mascaras, kajal sticks, eye shadows, nail varnishes and the like to which they also impart an interesting appearance. The percentage content of the microcapsules in these preparations can be from 0.1 to 90% by weight and is preferably from 1 to 50% by weight and more particularly from 2 to 10% by weight.

The cosmetic preparations may additionally contain mild, i.e. dermatologically compatible, **surfactants** of which examples are fatty alcohol polyglycol ether sulfates, monoglyceride sulfates, mono- and/or dialkylsulfosuccinates, fatty acid isethionates, fatty acid sarcosinates, fatty

acid taurides, fatty acid glutamates, α-olefin sulfonates, ether carboxylic acids, alkyl oligoglucosides, fatty acid glucamides, alkyl amidobetaines and/or protein fatty acid condensates (preferably based on wheat proteins).

The above-mentioned preparations may also contain other typical auxiliaries and additives which are largely identical with those already mentioned under the heading "active substances" and which therefore need not be listed again at this juncture. A number of formulations are shown in the Examples. Auxiliaries and additives which have not been mentioned thus far include the following:

Superfatting agents may be selected from such substances as, for example, lanolin and lecithin and also polyethoxylated or acylated lanolin and lecithin derivatives, polyol fatty acid esters, monoglycerides and fatty acid alkanolamides, the fatty acid alkanolamides also serving as foam stabilizers.

The **consistency factors** mainly used are fatty alcohols or hydroxyfatty alcohols containing 12 to 22 and preferably 16 to 18 carbon atoms and also partial glycerides, fatty acids or hydroxyfatty acids. A combination of these substances with alkyl oligoglucosides and/or fatty acid N-methyl glucamides of the same chain length and/or polyglycerol poly-12-hydroxystearates is preferably used.

Suitable **thickeners** are, for example, Aerosil® types (hydrophilic silicas), polysaccharides, more especially xanthan gum, guar guar, agar agar, alginates and tyloses, carboxymethyl cellulose and hydroxyethyl cellulose, also relatively high molecular weight polyethylene glycol mono- esters and diesters of fatty acids, polyacrylates (for example Carbopols® [Goodrich] or Synthalens® [Sigma]), polyacrylamides, polyvinyl alcohol and polyvinyl pyrrolidone, surfactants such as, for example, ethoxylated fatty acid glycerides, esters of fatty acids with polyols, for example pentaerythritol or trimethylol propane, narrow-range fatty alcohol ethoxylates or alkyl oligoglucosides and electrolytes, such as sodium chloride

and ammonium chloride.

Suitable **cationic polymers** are, for example, cationic cellulose derivatives such as, for example, the quaternized hydroxyethyl cellulose obtainable from Amerchol under the name of Polymer JR 400®, cationic starch, copolymers of diallyl ammonium salts and acrylamides, quaternized vinyl pyrrolidone/vinyl imidazole polymers such as, for example, Luviquat® (BASF), condensation products of polyglycols and amines, quaternized collagen polypeptides such as, for example, Lauryldimonium Hydroxypropyl Hydrolyzed Collagen (Lamequat® L, Grünau), quaternized wheat polypeptides, polyethyleneimine, cationic silicone polymers such as, for example, amodimethicone, copolymers of adipic acid and dimethylamino-hydroxypropyl diethylenetriamine (Cartaretine®, Sandoz), copolymers of acrylic acid with dimethyl diallyl ammonium chloride (Merquat® 550, Chemviron), polyaminopolyamides as described, for example, in **FR 2 252 840 A** and crosslinked water-soluble polymers thereof, cationic chitin derivatives such as, for example, quaternized chitosan, optionally in micro-crystalline distribution, condensation products of dihaloalkyls, for example dibromobutane, with bis-dialkylamines, for example bis-dimethylamino-1,3-propane, cationic guar gum such as, for example, Jaguar®CBS, Jaguar®C-17, Jaguar®C-16 of Celanese, quaternized ammonium salt polymers such as, for example, Mirapol® A-15, Mirapol® AD-1, Mirapol® AZ-1 of Miranol.

Suitable **anionic, zwitterionic, amphoteric and nonionic polymers** are, for example, vinyl acetate/crotonic acid copolymers, vinyl pyrrolidone/vinyl acrylate copolymers, vinyl acetate/butyl maleate/isobornyl acrylate copolymers, methyl vinylether/maleic anhydride copolymers and esters thereof, uncrosslinked and polyol-crosslinked polyacrylic acids, acrylamidopropyl trimethylammonium chloride/acrylate copolymers, octylacrylamide/methyl methacrylate/tert.-butylaminoethyl methacrylate/2-hydroxypropyl methacrylate copolymers, polyvinyl pyrrolidone, vinyl

pyrrolidone/vinyl acetate copolymers, vinyl pyrrolidone/dimethylaminoethyl methacrylate/vinyl caprolactam terpolymers and optionally derivatized cellulose ethers and silicones.

Suitable **silicone compounds** are, for example, dimethyl polysiloxanes, methylphenyl polysiloxanes, cyclic silicones and amino-, fatty acid-, alcohol-, polyether-, epoxy-, fluorine-, glycoside- and/or alkyl-modified silicone compounds which may be both liquid and resin-like at room temperature. Other suitable silicone compounds are simethicones which are mixtures of dimethicones with an average chain length of 200 to 300 dimethylsiloxane units and hydrogenated silicates. A detailed overview of suitable volatile silicones can be found in Todd et al. in **Cosm. Toil. 91, 27 (1976)**.

Typical examples of **fats** are glycerides while suitable **waxes** are inter alia natural waxes such as, for example, candelilla wax, carnauba wax, Japan wax, espartograss wax, cork wax, guaruma wax, rice oil wax, sugar cane wax, ouricury wax, montan wax, beeswax, shellac wax, spermaceti, lanolin (wool wax), uropygial fat, ceresine, ozocerite (earth wax), petrolatum, paraffin waxes, microwaxes; chemically modified waxes (hard waxes) such as, for example, montan ester waxes, sasol waxes, hydrogenated jojoba waxes and synthetic waxes such as, for example, polyalkylene waxes and polyethylene glycol waxes.

The total percentage content of auxiliaries and additives may be from 1 to 50% by weight and is preferably from 5 to 40% by weight, based on the particular formulation. The formulations may be produced by standard hot or cold processes and are preferably produced by the phase inversion temperature method. The decorative preparations typically contain microcapsules of only one type, for example containing pigments or care components. However, it is also possible to produce chitosan microcapsules charged with different substances. This includes on the one hand mixtures of microcapsules charged with different substances and on

the other hand microcapsules charged with one or more compatible substances.

Examples

Example H1. In a 500 ml three-necked flask equipped with a stirrer and reflux condenser, 3 g of agar agar were dissolved in 200 ml of water in boiling heat. First a homogeneous disperson of 10 g of glycerol and 2 g of talcum in 88 ml of water and then a preparation of 25 g of chitosan (Hydagen® DCMF, 1% by weight in glycolic acid, Henkel KGaA, Düssledorf/FRG), 10 g of paraffin oil, 0.5 g of Phenonip® (preservative mixture containing phenoxyethanol and parabens) and 0.5 g of Polysorbate-20 (Tween® 20, ICI) in 64 g of water were then added to the mixture over a period of about 30 mins. with vigorous stirring. The matrix obtained was filtered, heated to 60°C and added dropwise to a 0.5% by weight sodium alginate solution. To obtain microcapsules of the same diameter, the preparations were then sieved.

Example H2. In a 500 ml three-necked flask equipped with a stirrer and reflux condenser, 3 g of agar agar were dissolved in 200 ml of water in boiling heat. First a homogeneous disperson of 10 g of glycerol and 2 g of talcum in ad 100 g water and then a preparation of 25 g of chitosan (Hydagen® DCMF, 1% by weight in glycolic acid, Henkel KGaA, Düssledorf/FRG), 0.5 g of tocopherol acetate and 0.5 g of Phenonip® in ad 100 g water were added to the mixture over a period of about 30 mins. with vigorous stirring. The matrix obtained was filtered, heated to 50°C and dispersed with vigorous stirring in 2.5 times its volume of paraffin oil cooled beforehand to 15°C. The dispersion was then washed with an aqueous solution containing 1% by weight of sodium lauryl sulfate and 0.5% by weight of sodium alginate and then repeatedly with a 0.5% by weight

aqueous Phenonip solution, the oil phase being removed in the process. An aqueous preparation containing 8% by weight of microcapsules with a mean diameter of 1 mm was obtained after sieving.

Example H3. In a 500 ml three-necked flask equipped with a stirrer and reflux condenser, 3 g of agar agar were dissolved in 200 ml of water in boiling heat. First a homogeneous disperson of 10 g of glycerol and 2 g of talcum in 88 ml of water and then a preparation of 2.5 g of sodium alginate in the form of a 10% by weight aqueous solution, 1 g of iron oxide, 0.5 g of Phenonip® and 0.5 g of Polysorbate-20 (Tween® 20, ICI) in 64 g of water were added to the mixture over a period of about 30 mins. with vigorous stirring. The matrix obtained was filtered, heated to 60°C and added dropwise to a 1% by weight solution of chitosan glycolate in water. To obtain microcapsules of the same diameter, the preparations were then sieved.

Example H4. In a 500 ml three-necked flask equipped with a stirrer and reflux condenser, 3 g of agar agar were dissolved in 200 ml of water in boiling heat. First a homogeneous disperson of 10 g of glycerol and 2 g of talcum in ad 100 g water and then a preparation of 2.5 g of sodium alginate in the form of a 10% by weight aqueous solution, 5 g of mica pigments and 0.5 g of Phenonip® in ad 100 g water were added to the mixture over a period of about 30 mins. with vigorous stirring. The matrix obtained was filtered, heated to 50°C and dispersed with vigorous stirring in 2.5 times its volume of paraffin oil cooled beforehand to 15°C. The dispersion was then washed with an aqueous solution containing 1% by weight of sodium lauryl sulfate and 0.5% by weight of chitosan glycolate and then repeatedly with a 0.5% by weight aqueous Phenonip solution, the oil phase being removed in the process. An aqueous preparation containing 8% by weight of microcapsules with a mean diameter of 1 mm was obtained after sieving.

Example H5. In a 500 ml three-necked flask equipped with a stirrer and reflux condenser, 3 g of agar agar were dissolved in 200 ml of water in boiling heat. First a homogeneous disperson of 10 g of glycerol and 2 g of talcum in ad 100 g water and then a preparation of 2.5 g of sodium alginate in the form of a 10% by weight aqueous solution, 5 g of camphor and 0.5 g of Phenonip® in ad 100 g water were added to the mixture over a period of about 30 mins. with vigorous stirring. The matrix obtained was filtered, heated to 50°C and dispersed with vigorous stirring in 2.5 times its volume of paraffin oil cooled beforehand to 15°C. The dispersion was then washed with an aqueous solution containing 1% by weight of sodium lauryl sulfate and 0.5% by weight of chitosan glycolate and then repeatedly with a 0.5% by weight aqueous Phenonip solution, the oil phase being removed in the process. An aqueous preparation containing 8% by weight of micro-capsules with a mean diameter of 1 mm was obtained after sieving.

The following Table contains a number of Formulation Examples for various decorative cosmetic products using chitosan microcapsules charged with active substances. All quantities are in % by weight. In the Table, (1) is a tinted day cream, (2) a powder cream, (3) a compressed face powder, (4) a loose face powder, (5) rouge, (6) lip gloss, (7) a care lipstick, (8) a decorative lipstick, (9) an eyeliner, (10) a mascara, (11) a compressed eye shadow; (2) an eye shadow in emulsion form, (13) a pearl varnish and (14) a cream varnish.

Table 1

Formulation Examples - Decorative Cosmetics (Part 1)

Composition	1	2	3	4	5	6	7
Glyceryl Stearate	4.0	1.8	-	-	-	-	-
PEG-20 Sorbityl Laurate	1.0	0.7	-	-	-	-	-
Cetyl Alcohol	1.0	-	-	-	-	-	-
Lanolinyl Alcohol	-	4.0	-	-	3.0	45.0	3.0
Stearic acid	2.0	-	-	-	-	-	-
Isopropyl Stearate	-	-	2.0	-	-	-	-
Cetylstearyl Octanoate	5.0	-	-	-	-	-	-
Octyl Palmitate	3.0	-	-	-	-	-	-
Oleyl Oleate	-	3.2	-	-	-	-	-
Dimethyl Polysiloxane	-	-	-	-	2.5	10.0	-
Stearoxydimethicone	-	-	-	-	-	-	1.0
Beeswax	-	-	-	-	-	2.0	-
Carnauba wax	-	-	-	-	-	1.0	3.0
Candelilla wax	-	-	-	-	-	-	7.0
Ozocerite	-	-	-	-	-	2.0	-
Castor oil	-	-	-	-	-	15.0	82.5
Lanolin	-	-	-	-	-	-	10.0
Chitosan microcapsules of Example H1	7.0	7.5	2.0	-	2.5	5.0	-
Chitosan microcapsules of Example H2	-	-	-	-	-	-	2.0
Chitosan microcapsules of Example H3	-	-	-	0.5	-	-	-
Chitosan microcapsules of Example H4	-	-	-	11.0	-	-	-
D-panthenol	-	-	-	-	-	-	0.5
Xanthan Gum	0.4	-	-	-	-	-	-
Magnesium Stearate	-	-	3.0	-	-	-	-
Zinc Stearate	-	-	-	-	4.5	-	-
Carboxymethyl cellulose	-	0.3	-	-	-	-	-
Polybutylene	-	-	-	-	-	17.0	-
Talcum	4.0	9.0	30.0	65.5	17.5	-	-
Kaolin	-	-	-	5.0	-	-	-
Magnesium Silicate	5.0	0.3	33.0	15.0	22.0	-	-
Titanium dioxide	1.5	4.0	5.0	-	8.0	-	-
Iron oxide	0.8	1.1	2.0	-	4.0	-	-
Mica pigments	-	-	7.0	-	35.0	3.0	-
Propylene glycol	5.0	10.0	-	-	-	-	-
Triethanolamine	-	1.0	-	-	-	-	-
Perfume oil	0.5	0.4	0.5	1.0	0.5	-	0.4
Water	to 100						

Table 1: Formulation Examples - Decorative Cosmetics (Part 2)

Composition	8	9	10	11	12	13	14
Glyceryl Stearate	-	-	6.0	-	5.5	-	-
Lanolyl Alcohol	-	-	-	0.5	1.5	-	-
Oleth-10	-	-	-	-	1.5	-	-
Stearic acid	-	-	1.0	-	1.0	-	-
Paraffin oil	-	-	49.0	-	-	-	-
Isopropyl Stearate	-	-	-	2.0	3.0	-	-
Isostearyl Isostearate	12.0	-	-	-	-	-	-
Myristyl Lactate	6.0	-	-	-	-	-	-
Dimethyl polysiloxane	-	-	-	-	2.0	-	-
Bees wax	3.0	-	16.0	-	2.5	-	-
Carnauba wax	2.5	-	3.0	-	-	-	-
Candelilla wax	2.0	-	-	-	-	-	-
Ozocerite	2.0	-	-	-	-	-	-
Castor oil	44.5	-	1.5	-	-	-	-
Lanolin	5.0	-	-	-	-	-	-
Chitosan microcapsules of Example H1	2.0	-	-	3.5	3.0	-	-
Chitosan microcapsules of Example H2	0.5	-	-	-	-	-	-
Chitosan microcapsules of Example H4	6.0	7.0	12.0	10.0	8.0	-	-
Chitosan microcapsules of Example H5	-	-	-	-	-	1.5	1.5
PVP/hexadecane copolymer	-	-	3.0	-	-	-	-
Polyvinyl pyrrolidone	-	-	-	0.5	-	-	-
Zinc Stearate	-	-	-	2.0	-	-	-
Carboxymethyl cellulose	-	1.5	-	-	-	-	-
Talcum	-	-	-	36.0	-	-	-
Kaolin	-	-	-	5.0	-	-	-
Magnesium silicate	-	1.5	-	-	2.0	-	-
Stearalkonium hectorite	-	-	-	-	-	1.0	1.0
Titanium dioxide	2.0	-	-	5.0	2.0	0.6	1.0
Bismuth oxychloride	-	-	-	20.0	-	0.2	-
Triethanolamine shellac	-	2.0	-	-	-	-	-
Organic pigments	12.0	-	-	15.0	8.0	1.2	1.0
Methyl acetate	-	-	-	-	-	8.0	10.0
Ethyl acetate	-	-	-	-	-	8.0	18.0
Propyl acetate	-	-	-	-	-	12.0	16.0
Butyl acetate	-	-	-	-	-	25.0	23.0
Polyester resin	-	-	-	-	-	7.5	-
Nitrocellulose/isopropyl alcohol	-	-	-	-	-	21.0	19.0
Toluene sulfonamide/formaldehyde resin	-	-	-	-	-	9.0	11.0
Polyacrylate	-	-	-	-	-	-	1.5
Camphor	-	-	-	-	-	1.5	1.5
Dibutyl phthalate	-	-	-	-	-	5.0	5.0
Propylene glycol	-	5.0	2.0	-	5.0	-	-
Perfume oil	0.4	-	-	-	0.5	-	-
Water	to 100						

CLAIMS

1. Decorative cosmetic preparations containing chitosan microcapsules charged with active substances.

2. Preparations as claimed in claim 1, characterized in that they contain active substances selected from the group consisting of cosmetic oils, pearlizing waxes, stabilizers, biogenic agents, UV protection factors, antioxidants, preservatives, insect repellents, self-tanning agents, perfume oils and dyes.

3. Preparations as claimed in claims 1 and/or 2, characterized in that they contain chitosan microcapsules charged with active substances which are obtained by

(a) preparing a matrix from gel formers, chitosans and active substances and

(b) treating the matrix with aqueous solutions of anionic polymers.

4. Preparations as claimed in claims 1 and/or 2, characterized in that they contain chitosan microcapsules charged with active substances which are obtained by

(a) preparing a matrix from gel formers, chitosans and active substances,

(b) dispersing the matrix in an oil phase,

(c) treating the dispersed matrix with aqueous solutions of anionic polymers and removing the oil phase in the process.

5. Preparations as claimed in claims 1 and/or 2, characterized in that they contain chitosan microcapsules charged with active substances which are obtained by

(a) preparing a matrix from gel formers, anionic polymers and active principles and

(b) treating the matrix with aqueous chitosan solutions.

6. Preparations as claimed in claims 1 and/or 2, characterized in that they contain chitosan microcapsules charged with active substances which are obtained by

(a) preparing a matrix from gel formers, anionic polymers and active principles,

(b) dispersing the matrix in an oil phase,

(c) treating the dispersed matrix with aqueous chitosan solutions and removing the oil phase in the process.

7. Preparations as claimed in at least one of claims 1 to 6, characterized in that the microcapsules contain heteropolysaccharides or proteins as gel formers.

8. Preparations as claimed in at least one of claims 1 to 7, characterized in that the microcapsules contain chitosans with an average molecular weight of 10,000 to 500,000 or 800,000 to 1,200,000 dalton.

9. Preparations as claimed in at least one of claims 1 to 8, characterized in that the microcapsules contain salts of alginic acid or anionic chitosan derivatives as anionic polymers.

10. The use of microcapsules charged with active substances for the production of decorative cosmetic preparations.

(12) NACH DEM VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS (PCT) VERÖFFENTLICHTE INTERNATIONALE ANMELDUNG

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Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkurzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen.

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(54) **Title:** DECORATIVE COSMETIC PREPARATIONS CONTAINING CHITOSAN MICROCAPSULES CHARGED WITH ACTIVE INGREDIENTS

(54) **Bezeichnung:** DEKORATIVE KOSMETISCHE ZUBEREITUNGEN MIT WIRKSTOFFEN BELADENE CHITOSANKAPSELN ENTHALTEND

(57) **Abstract:** Disclosed are decorative cosmetic preparations containing chitosan microcapsules charged with active ingredients.

(57) **Zusammenfassung:** Vorgeschlagen werden dekorative kosmetische Zubereitungen, welche mit Wirkstoffen beladene Chitosanmikrokapseln enthalten.

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Patent and Trademark Office

DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number	C 2041 PCT/US
First Named Inventor	DeMORAGAS, Maria
COMPLETE IF KNOWN	
Application Number	10/049,961
Filing Date	07/19/2002
Group Art Unit	
Examiner Name	

As a below named inventor, I hereby declare that:
My residence, post office address, and citizenship are as stated below next to my name.
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

DECORATIVE COSMETIC PREPARATIONS CONTAINING CHITOSAN MICROCAPSULES CHARGED WITH ACTIVE INGREDIENTS

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 08/09/2000 as United States Application Number or PCT International Application Number PCT/EP00/07717 and was amended on (MM/DD/YYYY) _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY	Priority Not Claimed	Certified Copy Attached? YES	NO
99116262.9	EP	08/18/1999	☐	☐	☒

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)

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U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number *(if applicable)*
	PCT/EP00/07717	08/09/2000	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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100

DECLARATION

ADDITIONAL INVENTOR(S) Supplemental Sheet

200

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City		State		Zip		Country		Applicant Authority	

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Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country		Applicant Authority	

☐ Additional inventors are being named on supplemental sheet(s) attached hereto